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強レーザー場中の分子のalignment過程について理論的考察を行った。特に、楕円偏光をもつレーザー場の場合、非対称コマ分子の回転定数および分極率成分の非対称性が、分子の3つの主軸の空間フレームに対するalignmentの程度に、どのように反映するかを調べた。

1 Introduction

In an intense laser field, rotational motion of molecules is hindered by the interaction between the laser electric field and the induced dipole moment. The resultant quantum states are called pendular states. In the case of linear molecules interacting with a linearly polarized laser field, the axes of molecules in the pendular states are aligned along the laser polarization direction [1]. In an elliptically polarized laser field, it has been reported that all the three principal axes of an asymmetric top molecule can be aligned with respect to a given laboratory frame [2].

In the present study, the alignment of asymmetric top molecules in an elliptically polarized intense laser field is investigated theoretically in order to clarify the relationship between (i) the degree of the alignment of the three principal axes and (ii) that of the asymmetry in the rotational constants and the polarizability tensor components.

2 Model

The degree of the asymmetry in three rotational constants A, B, and C is characterized by Ray's asymmetry parameter; κ,

$$\kappa = (2B - A - C)/(A - C). \qquad (-1 \leq \kappa \leq 1) \qquad (1)$$

Similary, the degree of the asymmetry in the diagonal elements of a polarizability tensor is described by η defined as

$$\eta = (2\alpha_{bb} - \alpha_{aa} - \alpha_{cc})/(\alpha_{aa} - \alpha_{cc}). \quad (-1 \leq \eta \leq 1) \tag{2}$$

At the limit of a prolate symmetric top, $\kappa = \eta = -1$, and at the limit of an oblate symmetric top, $\kappa = \eta = 1$.

By changing systematically these asymmetry parameters while keeping the values of the maximum and minimum rotational constants A and C, and those for the polarizability components α_{aa} and α_{cc}, the quantum states for the rotational motion of asymmetric top molecules in the time-averaged interaction potential are obtained. The degree of the alignment of the three principal axes in the resultant states are represented as the expectation values of the square modulus of the direction cosines between the molecular principal axes and the space-fixed axes.

3 Results and Discussion

It is shown that the degree of alignment of three principal axes in the pendular states of an asymmetric top molecule is sensitively dependent on κ and η.

When η increases, the degree of alignment of the a axis decreases monotonically, while that of the c axis increases monotonically. The degree of alingment of the b axis is maximized at around $\eta = 0$. These tendencies are interpreted in terms of the variation of the pendular potential with respect to the asymmetry in the diagonal components of the polarizability tensor.

When κ increases, the degree of alignment of the respective principal axes decreases monotonically. This is interpreted by the increase in the energies of the pendular states associated with the decrease in the moment of inertia for the pendular motion, inducing the delocalization of the wavefunction of the pendular states.

References

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